Ordering, tunability, and stability in the formation of semiconductor quantum dots

R. Leon*‡, M. Stevens-Kalceff§, J. Zou#, C. Lobo‡ and D. J. H. Cockayne#

*Jet Propulsion Lab, California Institute of Technology, Pasadena, CA 91109-8099, U.S.A.
‡Australian National University, Canberra, ACT 0200, Australia.
§University of Technology, Sydney, P.O. Box 123, Broadway, NSW 2007, Australia
#Electron Microscopy Unit, The University of Sydney, NSW 2006, Australia

By varying growth conditions in Metalorganic Vapor Phase Epitaxy (MOVPE) and using several imaging and spectroscopic techniques [Force Microscopy (FM), Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM) plus Cathodoluminescence (CL) imaging and spectroscopy], different strategies to tune Stranski-Krastanow (S-K) III-V quantum dots were determined; conditions for both thermodynamical stability and for Ostwald ripening of the islands were realized; and also, two methods to induce one-dimensional alignment in InGaAs/GaAs quantum dots (QDs) were found.

Tunability in the concentration and average dimensions of self-forming semiconductor quantum dots (QD's) has been attained. Three of the approaches examined here are: variations with temperature, group V partial pressure and with substrate miscut angle. Thermally activated group III adatom mobilities result in larger diameters and lower concentrations with increasing deposition temperatures. These variations are presented for both InGaAs/GaAs [1] and AlInAs/AlGaAs [2] quantum dots, where striking differences were seen. Tunability in the InGaAs/GaAs QD concentration was also obtained in MOVPE by varying the arsine flow. The latter gave widely varying concentrations and similar sizes. Substrate orientation was found to also be a key factor in island nucleation: Changes in vicinal orientation near (100) can be used to exploit the preferential step edge nucleation at mono and multi-atomic steps, so varying miscut angle can be used to change island densities and sizes. Furthermore, a link between step separation and island uniformity was found. Step availability for a given adatom diffusion length was seen to be a major determinant of island uniformity at temperatures below activation for mobile clusters [3]. Anisotropies in island nucleation producing n-dot strings aligned with multi-atomic step edges were observed for miscuts $\geq$ 0.75°.

We also observed diverging behaviors in the S-K InGaAs/GaAs island formation: varying the group V partial pressures results in different onsets of the S-K transformation, island surface coverages, varying ratios between coherent and incoherent islands, and dissimilar morphologies upon annealing. The later experiments showed that island shape metastability and Ostwald ripening can be observed for some growth conditions; while high concentrations of smooth, lens shaped, small islands can also exist in thermodynamical equilibrium and do not ripen, if formed under conditions that minimize the InGaAs surface energy component.

An interesting morphological transformation was found in a multilayered InGaAs/GaAs quantum dot (QD) structure. Islands undergo a nucleation transition in between two types of step alignment after formation of the dislocation array. These are shown in figure 1. Surface probe microscopy also showed a transition in QD sizes and concentrations which shows in the CL spectroscopy in the form of a bimodal peak that can be de-convoluted into a high energy and a lower energy peak (shown in figure 2). The lower energy peak contribution becomes more significant when probing nearer the surface due to lower e- beam penetration depth. Plan view CL imaging and cross-sectional TEM showed that a dislocation array formed at the interface between GaAs and the first InGaAs QD layer. Strong QD luminescence was obtained despite the presence of the dislocation network.

References:

1. D. Leonard et al., *Appl. Phys. Lett.* **63**, 3203 (1993).
2. R. Leon, P. M. Petroff, D. Leonard, and S. Fafard, *Science* **267**, 1966 (1995)
3. R. Leon, T. J. Senden, Yong Kim, C. Jagadish, and A. Clark, *Phys. Rev. Lett* **78**, 4942 (1997).

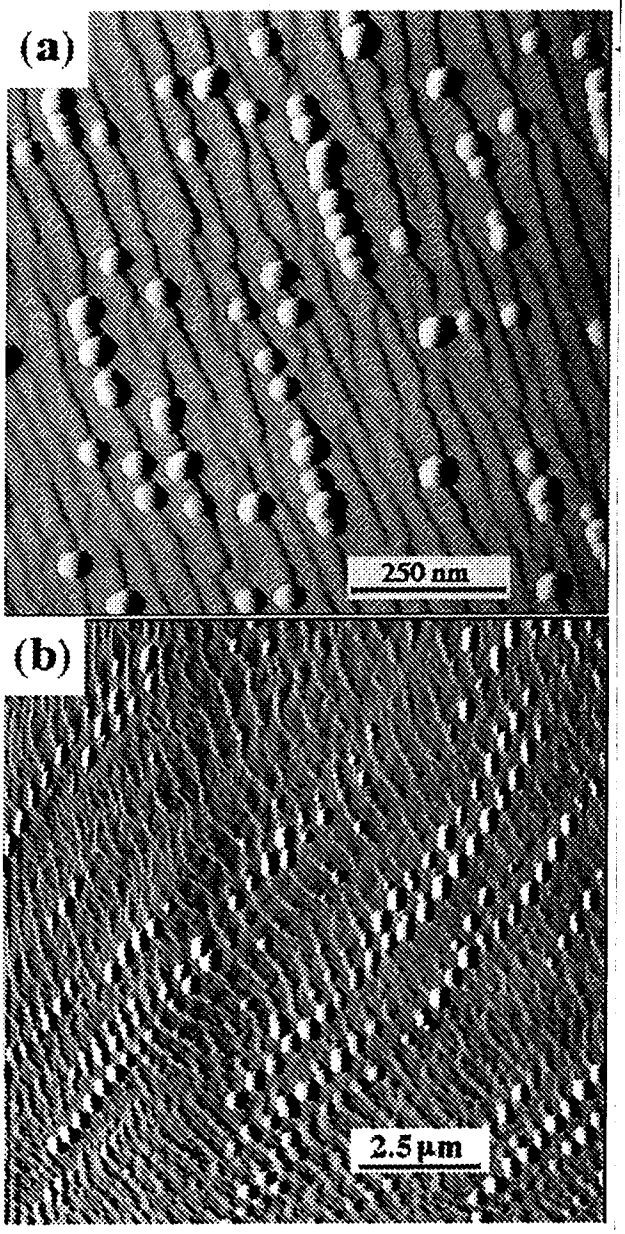

Figure 1 (left). Initial and final surface morphologies of a structure containing 100 layers of InGaAs QDs. Force Microscopy (FM) deflection images. (a) after the first layer deposition, (b) after 100 layers deposition, and (c) surface plot showing step alignment detail from (b).

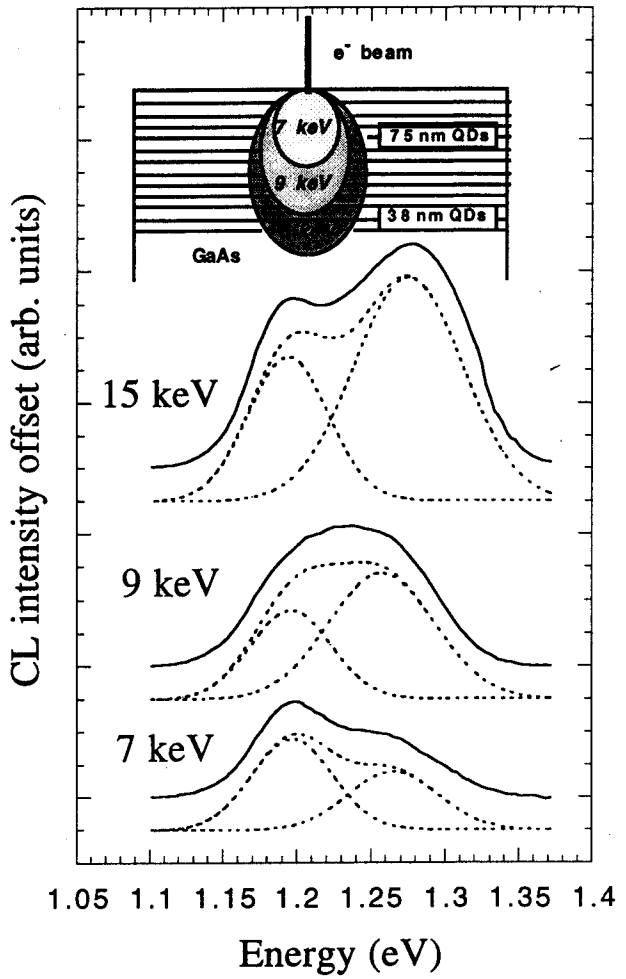

Figure 2 (above). CL spectra taken under different electron beam accelerating potentials. As can be seen in the spectra, the relative intensities of the two convoluted peaks changes with beam energy. Spectra have been fitted (shown in dotted lines) by adding two Gaussian peaks of varying intensities. The higher energy peak originates from the smaller QDs that are buried in the deeper layers [figure 1 (a)]. The contribution from the low energy peak is larger at 7 keV beam energy, which excites electron hole pairs closer to the surface, thereby, collecting a greater portion of the signal from the larger QDs. The relative intensities of the two convoluted peaks is seen to change with e beam penetration depth, and at 15 keV the high energy peak is of greater intensity. Integrated intensity ratios for the low energy/high energy peaks 0.65, 0.71 and 1.56 for 15 keV, 9 keV and 7 keV respectively. These correspond to calculated penetration depths of 1.22, 0.52 and 0.34 µm for the electron range in GaAs. Diagram on top of spectra shows a schematic of the electron beam penetration depth. The total thickness for the 100 layers of QDs is 1 µm.